

DDT in sediments and organisms around southern California outfalls

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CHLORINATED HYDROCARBONS are significant contaminants of the marine ecosystem off southern California (Figure 1). By far the most important of these synthetic organics in this region is DDT and its metabolites (total DDT). The dominant source of the DDT contamination has been identified as industrial wastes from one of the largest manufacturers of this pesticide. An undetermined but apparently very large quantity of these wastes was released to the sewer system of the County Sanitation Districts of Los Angeles County (CSDLAC) for up to two decades prior to 1970, when the input to the system was discovered and stopped.¹

Municipal and industrial wastewaters of this system undergo primary treatment at

the Joint Water Pollution Control Plant (JWPCP) before being discharged near Whites Point off Palos Verdes Peninsula. Unfortunately, reliable monitoring for DDT residues in JWPCP effluent did not begin until December 1970, after the release of these industrial wastes to the sewer system had ceased.² Despite source control, significant discharge of DDT compounds to the marine environment continued as the result of the heavy contamination of sediment deposits in the collection system.³ Here findings made during the last several years on major wastewater inputs of this contaminant, and on its spatial and temporal distributions in ocean bottom sediments, crabs, and flatfish collected near Los Angeles, are summarized.

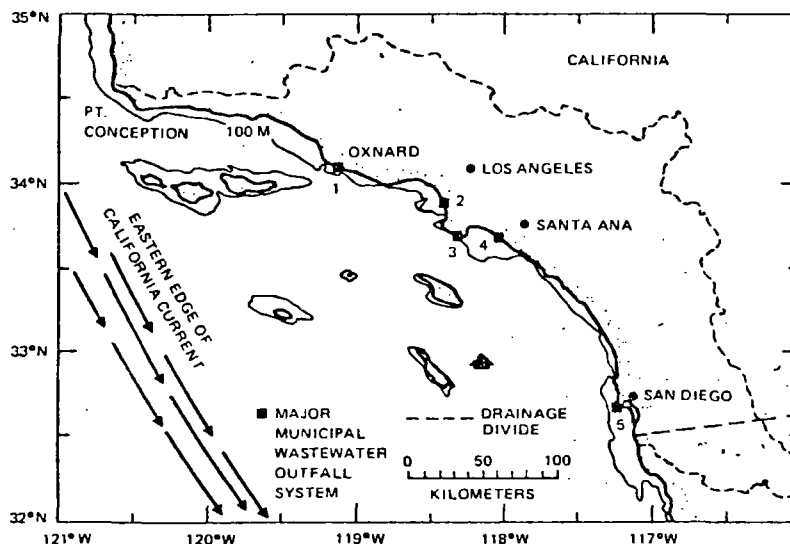


FIGURE 1.—The Southern California Bight. Outfall systems are: (1) Oxnard City, (2) Hyperion, Los Angeles City, (3) JWPCP, Los Angeles County, (4) Orange County, (5) Point Loma, San Diego City.

ANALYTICAL PROCEDURES

Wastewaters. Analyses of municipal wastewater concentrations of chlorinated hydrocarbons were conducted on 1-wk composites of final effluent, obtained from 3 to 24 daily grab samples; these samples were refrigerated until they could be composited proportionately according to wastewater flow rate at the end of the week. It was found that such storage prior to extraction causes less than 10 percent change in the individual composition of the chlorinated hydrocarbons in JWPCP final effluent. The following procedures were employed in the wastewater analyses conducted at this laboratory and at Bodega Marine Laboratory (BML) of the University of California at Berkeley.

Replicate composites approximately 1 l in volume were extracted in 2-l separatory funnels three times. The first solvent was 100 ml of 15 percent diethyl ether in hexane (by volume). The composite was shaken with the solvent for 2 min and the layers were allowed to separate; the organic solvent was then removed and placed in a round bottom flask. The aqueous fraction was extracted again with the second organic solvent, 100 ml of 6 percent diethyl ether in hexane, and the extract was added to the flask. The aqueous fraction was extracted a third time with 100 ml of hexane, and the extract was combined with the others in the flask. The extract was then reduced in volume and cleaned-up on the Florisil column described below. The procedure used at CSDLAC was very similar to the one just described, except that the solvent used first in the extraction was 24 percent diethyl ether in hexane.

Sediments. Approximately 5 g of the thawed sediment sample was dried on a steam table, weighed into a 250 ml Erlenmeyer flask with a standard taper joint fitted with a ground glass stopper, and covered with 50 ml of 24 percent (by volume) diethyl ether in hexane. A Florisil column was prepared as described below. The Erlenmeyer flask with the sediment and solvent was shaken, and the

solvent decanted onto the Florisil column. After the solvent passed through the Florisil column, the residue in the Erlenmeyer flask was suspended in a 50 ml volume of 6 percent diethyl ether in hexane; the flask was shaken and the solvent decanted onto the column. Following this, 50 ml of hexane were added to the flask, and the residue and solvent shaken again and decanted onto the Florisil column. The sample was then ready for injection into the gas chromatograph.

Organisms. Approximately 10 g of muscle tissue were dissected from thawed specimens of crabs and flatfish, homogenized in a small blender, and weighed into a beaker. The homogenate was covered with 20 ml of acetonitrile and the mixture ground with a high speed blender for about 1 min. The blade was rinsed twice with 20 ml of acetonitrile. This rinsing solution was added to the beaker with the sample, and then decanted and passed through filter paper into a 500 ml separatory funnel. The sample was rinsed 3 times with 20 ml of acetonitrile and the rinse decanted into the 500 ml separatory funnel with the other acetonitrile extract. Fifty ml of hexane were added to the 500-ml separatory funnel with the acetonitrile extract. The separatory funnel was shaken for a period of 1 min and the two layers were allowed to separate; the hexane (upper) layer was then collected. Extraction of the acetonitrile fraction was repeated twice with 50 ml of hexane, and the resulting hexane extracts were combined.

Florisil clean-up column. The hexane extracts were concentrated and cleaned up on a Florisil column. The Florisil (MCB, FX284) was activated at 700°C for 4 hr and stored under hexane in the dark until use. The Florisil was then packed in the column (30 mm op) to a height of 8 cm and covered with 1 cm of anhydrous granular sodium sulfate. After a sample was placed on the column, it was eluted with 45 ml of 6 percent diethyl ether in hexane (by volume).

Instrumentation. The samples were injected into a gas chromatograph with a ⁶³Ni electron-capture detector with the

TABLE I.—Average Concentrations ($\mu\text{g/l}$) and Mass Emission Rates (kg/yr) of the Isomers of Total DDT and its Metabolites

Year	Isomer						Total*
	p,p'-DDT	o,p'-DDT	p,p'-DDE	o,p'-DDE	p,p'-DDD	o,p'-DDD	
1971							
Avg. conc.	4.45	1.15	14.3	3.88	13.1	5.06	41.9
MER	2,280	590	7,340	1,990	6,720	2,600	21,500
1972							
Avg. conc.	0.73	0.22	3.96	0.25	6.54	1.54	13.2
MER	354	107	1,920	121	3,170	746	6,420
1973							
Avg. conc.	0.74	0.21	2.67	0.45	2.74	0.69	7.50
MER	367	104	1,320	223	1,360	342	3,720
1974							
Avg. conc.	0.10	0.03	1.41	0.06	1.17	0.25	3.02
MER	47.9	14.4	675	28.7	560	120	1,450

* Median annual total DDT concentrations for 1971 through 1974 are: 35.0, 12.4, 7.89, and 3.09 $\mu\text{g/l}$, respectively.

following parameters: nitrogen carrier gas with a flow rate of 80 ml/min; injector, column, and detector temperatures of 235°, 205°, and 280°C, respectively; glass columns, 6 mm o.d., 4 mm i.d., and 1.83 m (6 ft) long, packed with 5.5 percent OV-17 and 5.5 percent QF-1 on 80/100 mesh gas chrom Q. Calculations were made by comparison of sample peak heights to the peak heights of standards.

Similar instrumentation was employed at BML. However, at CSDLAC, a different gas chromatograph was used for the analysis. The column packing used was the same as that used by the SCCWRP laboratory.

WASTEWATER INPUTS

Independent measurements during 1971 and 1972 on split samples of jwpcp effluent at CSDLAC and BML yielded agreement on total DDT values within about 20 percent (R. Risebrough, BML, personal communication). Subsequent checks of CSDLAC monitoring data by BML and the authors' laboratory have confirmed this agreement. Table I summarizes the annual average concentrations and mass emission rates (MER) for DDT compounds discharged from the jwpcp outfall system between 1971 and 1974, calculated from monitoring data provided by CSDLAC. Corresponding estimated annual concentrations and MER's for the

four other major municipal outfall systems in the Bight during 1972 (the first year for which such estimates are available) were calculated using the results of BML analyses on single 1-wk composites of final effluent collected from each sewerage during the spring of 1972. These values are presented in Table II. These data indicate that more than 95 percent of the total DDT introduced to the coastal waters of the Bight via municipal wastewaters during 1972 was discharged via the jwpcp outfalls off Palos Verdes Peninsula.

Table III presents the median percent contribution of each isomer of DDT and its metabolites. These data indicate that the para-isomers of the DDD and DDE metabolites were the predominant contributors to the total DDT concentrations during 1971-72. The relative decrease in the percentage of DDT and the increase in the percentage of the DDD and DDE metabolites in jwpcp final effluent over the last 4 yr is probably the result of increased metabolization of DDT in the sewer lines without replenishment by fresh DDT wastes.

BOTTOM SEDIMENTS

In light of the more than 20 metric tons of total DDT which were discharged via jwpcp during 1971, and the unknown magnitude of such discharges in previous years,

TABLE II.—Concentrations ($\mu\text{g/l}$) and Estimated Mass Emission Rates (kg/yr) of the Isomers of Total DDT and its Metabolites

Discharger	Isomer						Total DDT
	p,p'-DDT	o,p'-DDT	p,p'-DDE	o,p'-DDE	p,p'-DDD	o,p'-DDD	
Hyperion-5 mi							
Concentrations	0.16	ND*	0.05	ND	ND	ND	0.21
MER	72.9	—	22.8	—	—	—	95.7
Hyperion-7 mi							
Concentrations	ND	ND	5.00	ND	ND	ND	5.00
MER	—	—	31.7	—	—	—	31.7
Orange County							
Concentrations	0.20	ND	0.16	ND	ND	ND	0.36
MER	41.4	—	33.1	—	—	—	74.5
Point Loma							
Concentrations	0.18	ND	0.08	ND	ND	ND	0.26
MER	23.6	—	10.5	—	—	—	34.1
Oxnard							
Concentrations	0.12	ND	0.04	ND	ND	ND	0.16
MER	1.66	—	0.55	—	—	—	2.21

* Not detected.

a detailed investigation was conducted into the quantity of DDT wastes contained in the bottom sediments of the JWPCP monitoring zone on the Palos Verdes shelf.⁴ Initially, during July 1971, box cores were collected at 4 stations (B18-B21) lying approximately at the discharge depth (60 m) and to the northwest of the outfall diffusers (Figure 2). In addition to measuring surface sediment concentrations, at the 4 sites, a vertical profile of total DDT was obtained at Station B21, where the highest surface concentration was found. Samples from this core were also analyzed independently by V. McClure of the National Marine Fisheries Service (La Jolla, Calif.). The results indicated agreement within about 20 percent over the entire range of concentrations measured (0.2–200

mg/dry kg). In June 1972, gravity cores were collected at 19 stations in the CSDLAC outfall monitoring grid (Region A). The following month, 4 additional gravity cores were collected at deeper sites off the outfalls to extend the study area (Region B). The 1972 cores were sectioned at 1 or 2 cm intervals to depths of 30 cm whenever possible, and at greater intervals thereafter. A total of 300 samples were analyzed for identifiable chlorinated hydrocarbons.

Figure 2 shows resultant surface sediment concentrations; the highest values occurred to the northwest of the outfalls, at approximately the depth of discharge. Similar contamination patterns for a number of trace metals have previously been reported.⁵ Table IV presents the median total DDT concentration as a function of depth for all the gravity cores collected in Region A during June 1972, and also the vertical profile obtained at Station 6B (0.2 km southwest of B21) in the area containing the highest concentrations. These results indicate that total DDT values typically decreased by a factor of 100 over a depth interval of about 24 cm in the outfall monitoring zone; however, in the region of greatest contamination, sediment concentrations at this depth were approxi-

TABLE III.—Annual Median Percent Contribution of Each Isomer of DDT and its Metabolites to the Total DDT Concentration

Isomers	1971	1972	1973	1974
p,p'-DDT	6.2	4.6	5.2	1.6
o,p'-DDT	2.5	1.5	2.8	0.7
p,p'-DDE	35	31	37	44
o,p'-DDE	8.5	1.9	3.8	0.4
p,p'-DDD	34	50	38	40
o,p'-DDD	11	12	9.1	7.5

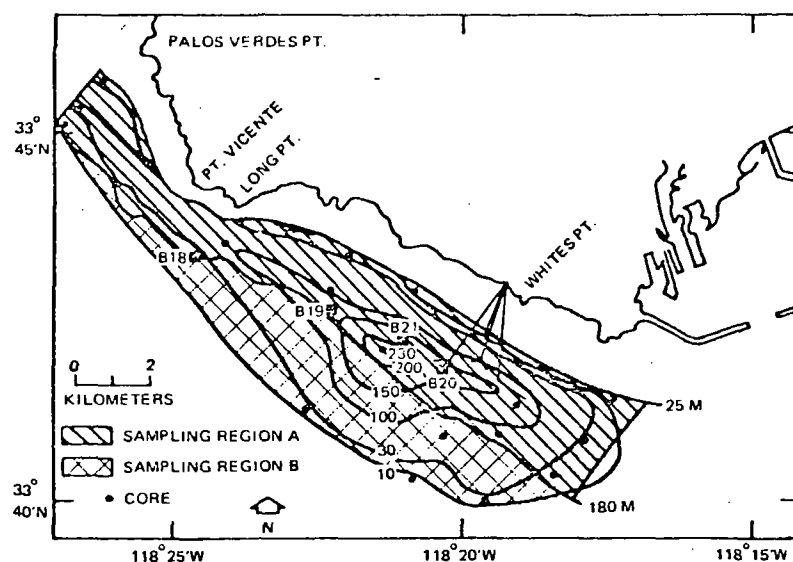


FIGURE 2.—Total DDT concentrations (mg/dry kg) in surface sediments (0 to 2 cm) off Palos Verdes, June 1972.

mately 10,000 times below the maximum surface concentration of 230 mg/dry kg.

The concentrations were contoured at each 2-cm interval down to 30 cm, and the quantity of total DDT in each interval of Regions A and B calculated, using a sediment density curve for these sediments derived from results obtained by Myers.⁶ The results are listed in Table V. These results show that in 1972, a very large quantity (> 150 metric tons) of DDT wastes was contained in the near-surface

sediments of the Palos Verdes shelf. Because the annual discharge rate of total DDT from the JWPCP outfalls decreased by 83 percent between 1971 and 1973, an attempt was made to determine whether there was any indication of large changes in sediment contamination levels over the 2-yr interval. In the summer of 1973, samples of surface sediments (0 to 5 cm)

TABLE IV.—Median Total DDT Concentrations (mg/dry kg)

Depth (cm)	Median Value of all June 1972 Short Gravity Cores	Core from Station 6B
0-2	26	230
2-4	21	150
4-6	23	270
6-8	8.4	180
8-10	4.2	97
10-12	8.8	88
12-14	2.1	44
14-16	2.0	2.7
16-18	0.7	0.1
18-20	0.1	0.2
20-22	0.5	0.1
22-24	0.2	0.03
24-26	0.3	0.02

TABLE V.—Total DDT (metric tons) in Palos Verdes Bottom Sediments, 1972*

Depth (cm)	Region A (28 sq km)	Region B (20 sq km)	Total (48 sq km)
0-2	8.7	6.9	15.6
2-4	9.7	5.4	15.1
4-6	10.1	7.5	17.6
6-8	13.4	6.4	19.8
8-10	16.7	6.7	23.4
10-12	14.3	6.7	21.0
12-14	6.4	2.8	9.2
14-16	7.8	3.7	11.5
16-18	8.4	4.7	13.1
18-20	2.0	0.9	2.9
20-22	2.5	1.2	3.7
22-24	1.2	0.4	1.6
24-26	0.4	0.2	0.6
26-28	0.1	0.05	0.15
28-30	0.1	0.05	0.15
Total	102	54	156

* Regions are shown on Figure 2.

TABLE VI.—Comparison of Total DDT Concentrations (mg/dry kg) in Surface Sediments

Sampling Location*	1971 (Box Core)	1972 (Gravity Core)	1973 (Ship's Grab)	1973 /1971
B18	29	19	26	0.9
B19	220	140	165	0.8
B20	330	170	290	0.9
B21	180	160	280	1.6
			Average	1.0
			Median	0.9

* Sampling locations are shown on Figure 2.

were collected by CSDLAC using a grab sampler, and analyzed for chlorinated hydrocarbons as part of their outfall monitoring program. Using these data, results obtained for the 3 yr at or near the original four box core stations sampled during 1971 were compared (Table VI). These data show considerable variation in the results. However, the average ratio of 1973:1971 total DDT concentrations for the four sites is 1.0 (median = 0.9), and there is no indication of a large decrease in the level of contamination of bottom sediments in the

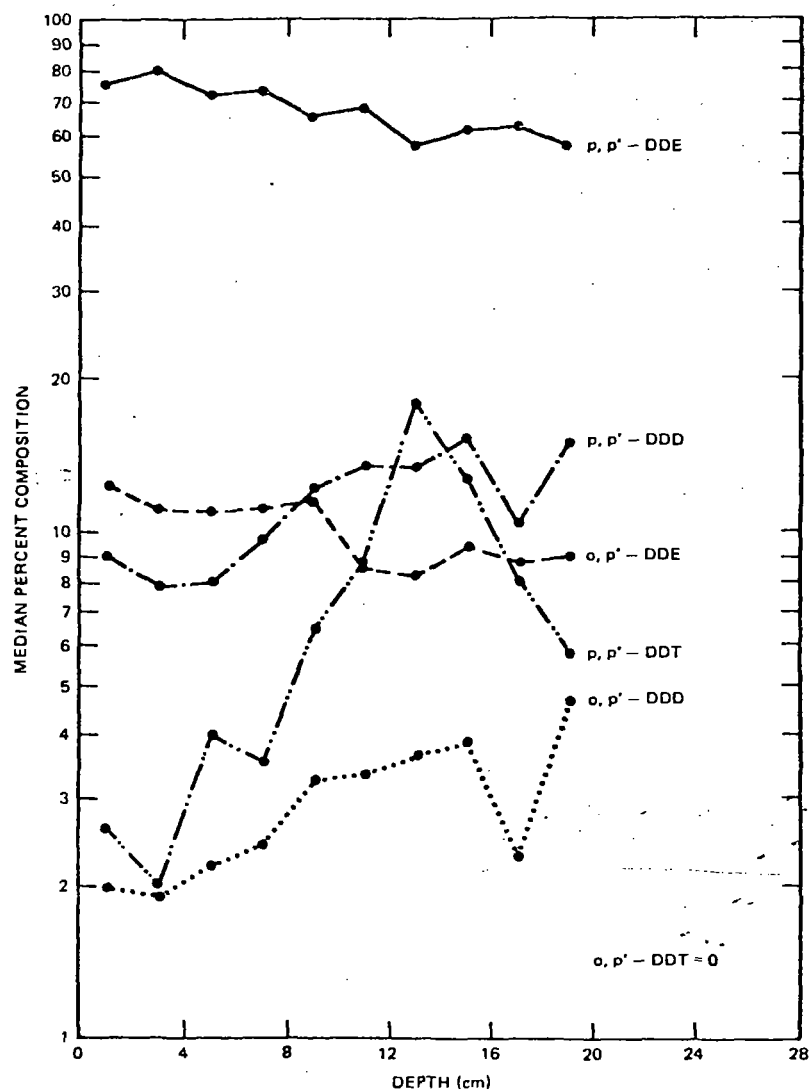


FIGURE 3.—Median percent contribution of each isomer of DDT and its metabolites to the "total DDT" value at each depth, short gravity cores, June 1972.

discharge area, despite the fact that the input rate from the outfalls had decreased by an order of magnitude. These results also show that at the four stations, the gravity cores collected in 1972 yielded considerably lower surface sediment concentrations than either the 1971 box cores or the 1973 grab samples. On the average, the 1972 values seemed to be low by about a factor of 1.5. To investigate this discrepancy further, Region A was contoured using the 1973 concentrations; the quantity of total DDT contained in the upper 5 cm was then calculated. The result was 37 metric tons, or 1.6 times the 23 metric tons obtained using the 1972 gravity core values for the same area and volume. Therefore, it appears that the 1972 inventory was probably somewhat low, and that approximately 200 metric tons of total DDT were contained in the bottom sediments of the Palos Verdes shelf.

As seen from the data of Figure 3, the major component found in the sedimentary column was p,p'-DDE. Although it constituted only 30 to 35 percent of the DDT dis-

TABLE VII.—Median Concentrations of Total DDT (mg/dry kg) in Surface Sediments

Outfall System	Core Depth (cm)	Concentration		
		Median	Range	n
JWPCP	0-2	26	2.8 -230	19
Hyperion:				
5 Mile	0-2	0.24	0.05 -0.56	21
7 Mile	0-2	0.66	0.22 -4.0	17
Orange Co.	0-3	0.07	0.03 -0.11	13
Point Loma	0-2	0.008	0.002-0.016	11
Oxnard	0-5	0.04	0.02 -0.10	9

charged from the JWPCP outfalls during 1971 and 1972, approximately 75 percent of the total DDT in the upper 5 cm of sediments and about 60 percent below 10 cm was p,p'-DDE. Only 2 to 3 percent of the total DDT found in the surface sediments in 1972 was p,p'-DDT. However, this percentage apparently was higher in the past; at a depth of 14 cm, p,p'-DDT comprised more than 15 percent of all the DDT compounds measured. The low level of DDT, relative to its metabolites, in the surface sediments

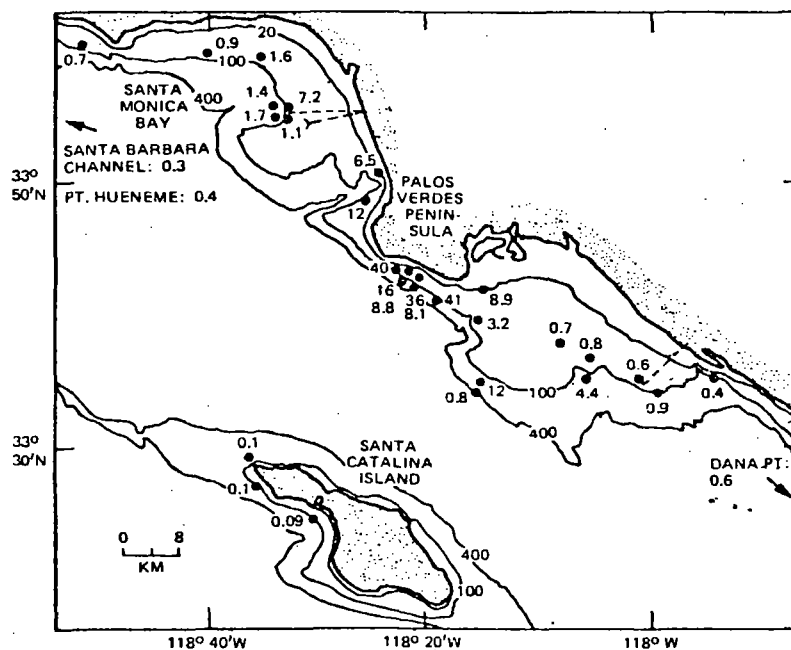


FIGURE 4.—Total DDT concentrations (mg/wet kg) in muscle tissue of Dover sole, *Microstomus pacificus*, collected off southern California, 1971-72.

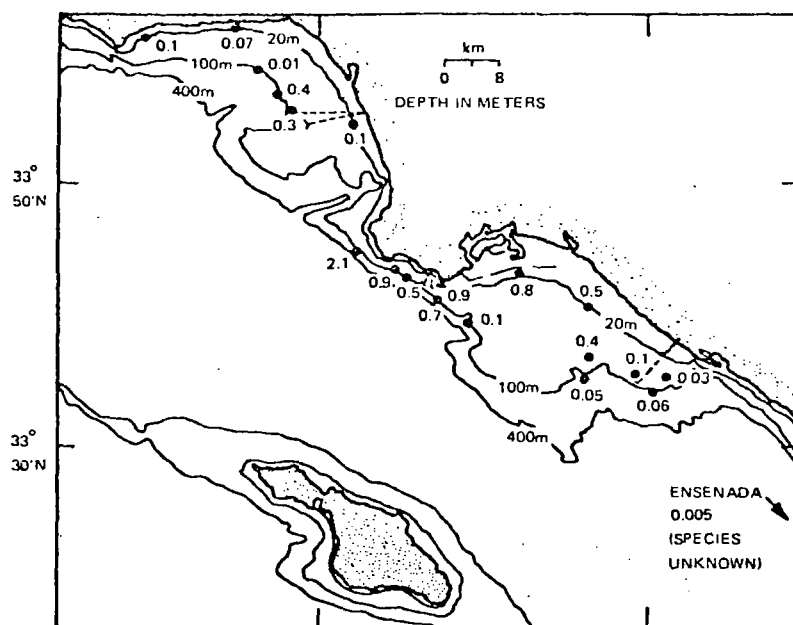


FIGURE 5.—Total DDT concentrations (mg/wet kg) in muscle tissue of the crab, *Cancer anthonyi*, collected off southern California, 1971-72.

is probably the result of its continued metabolism without comparable replenishment.

Differences between levels of DDT contamination on the Palos Verdes shelf and around other submarine outfall systems in the Bight are shown in Table VII. This table presents the median concentration of total DDT, as well as the range and the number of samples analyzed, in the surface layer of bottom sediments at each discharge site. Clearly, the benthic community off Palos Verdes Peninsula has been

exposed to levels of this contamination in the substrate which are several orders of magnitude greater than those found in the other outfall regions.

BENTHIC ORGANISMS

These regional gradients in sediment concentrations of DDT compounds were reflected in levels of this contaminant which were measured in flatfish and benthic crabs obtained at various stations in the Bight. Figures 4 and 5, respectively, illustrate total DDT concentrations found in muscle tissue of the Dover sole (*Microstomus pacificus*) and the yellow crab (*Cancer anthonyi*) collected during 1971-72. Both distributions exhibit concentrations in Palos Verdes specimens that are at least 10 to 100 times those measured in island or coastal controls. Table VIII presents median percentage compositions of the total DDT in the flatfish collected off Palos Verdes Peninsula. As was the case for the sediments, p,p'-DDE was the dominant form measured.

The U. S. Food and Drug Administration has established a limit of 5 ppm

TABLE VIII.—Median Percent Contribution of each Isomer of DDT and its Metabolites to the Total DDT Concentration in Muscle Tissue of Dover sole

Isomer	Median	Range	n
p,p'-DDT	0.7	ND*-1.4	18
o,p'-DDT	0.4	0.2-1.4	18
p,p'-DDE	84	77-90	18
o,p'-DDE	9.0	4.0-11	18
p,p'-DDD	6.1	3.4-11	18
o,p'-DDD	0.2	ND-0.8	18

* Not detected.

(mg/wet kg) for total DDT in seafood intended for interstate commerce. Although none of the crabs analyzed exceeded this value in the muscle tissue, more than two-thirds of the Dover sole collected from the Palos Verdes shelf during 1971-1972 did. Such excessive concentrations were not limited to Dover sole. The CSDLAC monitoring program has included occasional analyses of muscle tissue from two popular nearshore sportfish which feed on or near the bottom; these are the black perch (*Embiotoca jacksoni*) and the kelp bass (*Pseudopleuronectes clathratus*). During 1972, approximately 76 percent ($n = 41$) and 47 percent ($n = 19$) of these fish also exceeded the 5 ppm federal limit, respectively. Corresponding values for 1973 were 65 ($n = 60$) and 46 ($n = 28$) percent, respectively. (D. Hotchkiss, CSDLAC, personal communication).

Despite the fact that the annual emission of total DDT by the JWPCP outfalls decreased from nearly 22 metric tons in 1971 to less than 2 metric tons in 1974, no significant decrease in contamination of Dover sole trawled from the discharge region over approximately the same interval was found. Representative station values were obtained by taking the median of all analyses on fish obtained from trawls, at each location to eliminate the potential bias of different sample sizes from the individual stations off Palos Verdes. The median of these representative station values for the concentration of total DDT in the muscle tissue of the 1971-1972 Palos Verdes specimens was 9.0 mg/wet kg, with a range of 3.2 to 45 mg/wet kg ($n = 31$). Corresponding station values during 1974-1975 had a median DDT level of 12 mg/wet kg, with a range of 9.7 to 25 mg/wet kg ($n = 35$). Application of the non-parametric Wilcoxon signed-rank test to these data indicated no statistically significant difference at the 95 percent confidence level.

Thus, the protracted contamination of benthic fish in this region appears to be due largely to the very high levels of DDT compounds contained in the bottom sediments of the shelf. It is important that

monitoring of these chlorinated hydrocarbons be continued off Palos Verdes and throughout the Bight, to determine the persistence of this type of synthetic contaminant at a coastal discharge site, and its rate of dispersion to adjoining regions.

SUMMARY

The marine ecosystem off southern California is still highly contaminated by DDT compounds. The major source of this contamination appears to have been industrial wastes discharged in Los Angeles County municipal wastewater, which accounts for more than 95 percent of the total DDT measured in municipal wastewaters released to this ecosystem. As a result, approximately 200 metric tons of these chlorinated hydrocarbons have accumulated in bottom sediments within a 50 sq km area of the Palos Verdes shelf. Of this total, 70 percent was contained in the upper 12 cm in 1972, and surface layer concentrations exceeded 200 mg/dry kg near the JWPCP outfall diffusers. Concentrations of total DDT typically fell by a factor of 100 at a sediment depth of 20 cm in the monitoring region, and the median surface sediment concentration was 40 to 3,200 times those measured around the other four major submarine outfall systems. Seventy-five percent of the total DDT on the surface of the shelf was p,p' -DDT, and less than 3 percent was p,p' -DDT; however, the latter isomer constituted 15 percent of the total at a depth of 14 cm.

Benthic crabs and flatfish also showed very large contamination gradients; Palos Verdes specimens exceeded island or coastal control total DDT levels by a factor of 100. As with the surface sediments, the principal compound measured in the muscle tissue of these organisms was p,p' -DDT. Although none of the crabs collected in 1971-72 contained more than 5 ppm (mg/wet kg) total DDT, approximately 70 percent of the Dover sole exceeded this federal limit at that time.

Despite the fact that the discharge of total DDT via the JWPCP outfalls dropped from almost 22 metric tons in 1971 to less than 2 metric tons in 1974, little change

has been observed in surface sediment concentrations on the Palos Verdes shelf. In addition, Dover sole from this region showed no significant decreases over a 3-yr interval; median muscle tissue concentrations in 1971-1972 and 1974-1975 were 9.0 and 12 ng/wet kg respectively. Thus, it appears that the sediment reservoir of DDT wastes off Palos Verdes Peninsula now may be the dominant source of this contaminant to the benthic organisms in this region of the coastal ecosystem.

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